ELSEVIER International Journal of Pharmaceutics 144 (1996) 187–194

international journal of pharmaceutics

The influence of particle size and shape on the angle of internal friction and the flow factor of unlubricated and lubricated powders

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Received 11 July 1996; accepted 10 September 1996

Abstract

The shear properties of 8 different powders, which varied in particle size and shape, were studied using an annular shear cell. The angle of internal friction and the flow factor were determined. Magnesium stearate was added in concentrations between 0.25 and 1.25% w/w, and the change in the shear properties was recorded. For the unlubricated powders, the angle of internal friction was found to depend both on particle size and shape in a nonlinear manner, whereas the flow factor depended only on particle shape. The optimal magnesium stearate content, i.e. the concentration which gave the lowest angle of internal friction, varied for the powders. Both the angle of internal friction and the corresponding concentration of magnesium stearate depended only on particle shape. A large value for the aspect ratio as obtained for needle shaped particles was accompanied by a particularly high angle of internal friction. The optimal magnesium stearate concentration was least for needle shaped or round particles. The optimal magnesium stearate contents for the flow factor, i.e. the concentrations which gave the highest values of flow factor, were partly different from those obtained for the angle of internal friction. While the flow factor depended only on particle shape, the corresponding optimal magnesium stearate concentration was found to depend only on the particle size. For powders of comparable chemical composition such as maize starch and starch 1500, or microcrystalline and microfine cellulose, the angle of internal friction at an optimal lubricant concentration was found to be proportional to the elastic properties of the powders. Copyright © 1996 Elsevier Science B.V.

Keywords: Angle of internal friction; Annular shear cell; Flow factor; Particle shape; Particle size

1. Introduction

The flowability of powders determines their handling properties with respect to the manufac-

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PII S0378-5173(96)04755-2

ture of a variety of solid dosage forms, and a poor powder flow often requires major changes of an otherwise satisfactory formulation.

The influence of particle size and shape in terms of powder packing has often been described (Shotten and Obiorah, 1973; Ridgway and Morland, 1977; Lam and Nakagawa, 1994; Podczeck and Sharma, 1996). However, no general rule, applicable for a variety of shapes and size distributions, has been established.

Magnesium stearate is the most frequently used additive to improve the flow properties of powders. It has been proposed that magnesium stearate reduces the adhesion due to long-range van der Waals forces between the particles of a powder bed (Gold et al., 1968). Jones (1968) doubted this opinion because of the hydrophobic nature of magnesium stearate. However, in the adhesion literature, hydrophobization of materials has been reported to reduce the forces of adhesion significantly (Zimon, 1982; Deryaguin et al., 1978). An optimal magnesium stearate content, i.e. the concentration which improved powder flow most, can be found when a complete film has been formed surrounding each individual particle. However, above the optimal concentration, i.e. when the film formed increases in thickness, or when an overshot of fine particles exists, there is a sharp drop in flowability (Jones and Pilpel, 1966; Gold et al., 1968; Irono and Pilpel, 1982). This cannot only be attributed to a weakening of attractive forces between the host particles. During powder flow, particles are in frictional contact with each other. The three basic elements of friction are (i) the area of true contact between the sliding particles, (ii) the type and strength of the attractive forces between the contacting surfaces, and (iii) the shearing and rupture of the materials at the contact points and the surrounding area during sliding (Tabor, 1981). For lubricants such as magnesium stearate, which form a film around the particles, the last point is of major importance. The coefficient of friction μ between two materials is defined as the ratio between the shear strength of the formed adhesive junctions s and the yield pressure p_y of the softer material ($\mu = s/p_y$; Bowden and Tabor, 1964). In the absence of a lubricant, the values of s and p_y are those characteristic for the unlubricated powder, and the ratio between the shear stress τ_1 applied and the normal pressure P_1 at the contact points of the surface asperities, α_1, equals μ ($\alpha_1 = \tau_1/P_1 = \mu$; see Halling, 1981). If a lubricant film is present, the ratio between the shear stress τ_2 and the normal pressure P_2 at the contact area between lubricant film and asperities of the powder particles, α_2, determines the friction properties of the powder bed. As long as $\alpha_1 \geq \alpha_2$, the interparticulate friction will decrease with increasing film thickness. However, the value of α_1 is a constant, and the value of α_2 increases proportionally to the film thickness, because the shear stress applied is constant, but the normal pressure is reduced due to the increasing distance between the surface of the film, where shearing occurs, and the surfaces of the particles (Halling, 1981). Thus, as soon as the value of α_2 becomes larger than the constant value of α_1, a further increase in film thickness will result in increased friction and thus a decrease in powder flow. Consequently, the optimal magnesium stearate concentration is found at the minimum, if any friction characteristics (for example the angle of internal friction obtained from shear cell measurements) or the powder flowability is plotted as a function of the magnesium stearate concentration.

The aim of the present study was to relate the optimal magnesium stearate concentration of different host powders to variations in particle shape and size. Primarily, the powders were chosen to provide round, angular, cubic or needle shape according to BP. For each class of particle shape, a fine and a coarse powder were used, where fine is here defined for Edmundson's weight mean of the surface distribution d_{vs} (see Martin et al., 1987) to be at least 5 times smaller than d_{vs} of the coarse powder of similar shape. The particle shape was further classified in terms of the particle aspect ratio and the geometric shape.

2. Materials and methods

2.1. Materials

Acetylsalicylic acid (Rhone Poulenc, Dagen-

Acetylsalicylic acid (Rhone Poulenc, Dagenham, UK), maize starch (Roquette, Lestrem, France), microcrystalline cellulose (Emcocel®, Edward Mendell, NY), microfine cellulose (Elcema G250®, Rettenmaier, Ellwangen-Holzmühle, Germany), paracetamol (Becpharm, Essex, UK), potassium chloride, precipitated calcium carbonate (BDH, Poole, UK), and pregelatinized starch (Starch 1500®, Colorcon, Orpington, UK) were used as host powders. Magnesium stearate (BDH, Poole, UK) was used as lubricant/glidant.

2.2. Methods

The particle size and shape of the powders were assessed using an image analyzer (Seescan Solitaire 512, Cambridge, UK) attached to a microscope (Olympus BH-2, Tokyo, Japan) via a black/white CCD-4 camera (Rengo, Toyohashi, Japan). The powders were suspended in diiodomethane (Aldrich Chemicals, Fillingham, UK), which was found to separate any agglomeration due to its nonpolar character and its low surface free energy, and which provided sharp images due to its higher refractive index. For each powder, particle size and shape were measured simultaneously for 512 particles (maximum storage capacity of the image analyzer).

Powder mixtures of 0.25, 0.50, 0.75, 1.00 or 1.25% w/w magnesium stearate and host powder were prepared in a turbula mixer (Type Schatz T2C, Willy A. Bachofen, Basel, Switzerland) at a speed of 62 rpm. The mixing time was 3 min, and the batch size was 200 g.

The annular shear cell used (Technigraph, Bristol, UK) is similar to that described by Carr and Walker (1970). The maximum shear stress was measured by a load transducer (Ether UF2, Pioden Controls, Canterbury, UK) and recorded with a chart recorder (Servogor 120, BBC Goerz Metrawatt, Vienna, Austria). Shearing of a powder commenced at a chosen consolidation load, and the torque on the lid was monitored. When a stable reading had been obtained, the direction of the turntable was reversed, and the powder bed was reconsolidated to the initial failure stress. The measurements were repeated under subsequently reduced normal loads. Three different consolidation loads were applied, always using fresh powder. The results are reported as angle of internal friction, δ, and Jenike's flow factor, *ff* (Jenike, 1961). Based on *ff*, powders can be classified as 'cohesive non-flowing' ($ff < 2$), 'cohesive' (ff 2– < 4), 'ready flowing' (ff 4–10), and 'freely flowing' ($ff > 10$) (Rumpf, 1990).

Statistical analysis of the data was performed using commercial software (SPSS version 4.0, SPSS, UK).

3. Results and discussion

Magnesium stearate can act as a lubricant or glidant, if added to a powder. In this paper, magnesium stearate is regarded as a lubricant with respect to its ability to reduce friction, e.g. when discussing the values obtained for the angle of internal friction. On the other hand, magnesium stearate will be classified as a glidant when discussing the effects on powder flow, represented by Jenike's flow factor.

Table 1 summarizes the particle size and shape of the unlubricated powders. For the determination of Edmundson's weight mean of the surface distribution, d_{vs}, 36 Feret diameters have been measured across each particle at an interval of 10°, and the mean Feret diameter was calculated per particle. The value of d_{vs} was then calculated from the particle size distribution obtained. For needle shaped particles it would be better to use the length of the particles instead of d_{vs}. For microcrystalline cellulose the average length found was 80.7 μm, and for acetylsalicylic acid this value was 784.4 μm. In the following analyses, however, d_{vs} was used, because the length values are very similar to the d_{vs} values of the two powders. The aspect ratio, AR, is here defined as the ratio between largest Feret diameter and Feret diameter perpendicular to the largest Feret diameter (see Schneiderhöhn, 1954). The geometric shape has been assessed as shape factor SF (Podczeck, 1997) from the number of sharp corners, the particle elongation, and the deviations from the shapes of a triangle, circle and square:

Table 1
Particle size and shape of powders

Powder	d_{vs} [μm]	Shape (BP)	AR	SF
Pregelatinized starch	103.2	Angular	1.38 ± 0.26	7.54 ± 0.35
Paracetamol	537.6	Angular	1.61 ± 0.65	7.38 ± 0.53
Calcium carbonate	4.6	Cubic	1.20 ± 0.20	7.66 ± 0.36
Potassium chloride	481.1	Cubic	1.27 ± 0.29	7.70 ± 0.56
Maize starch	49.2	Round	1.16 ± 0.12	3.86 ± 0.18
Microfine cellulose	363.3	Round	1.40 ± 0.36	4.48 ± 0.78
Microcryst. cellulose	107.7	Rod shaped	2.19 ± 0.99	7.16 ± 0.43
Acetylsalicylic acid	721.7	Needle shaped	3.47 ± 1.17	7.45 ± 0.39

d_{vs}, Edmundson's weight mean of the surface distribution.

$$SF = Co + \frac{P}{l} \times \frac{A}{\frac{s \times s_p}{2}} - \frac{A}{\frac{\pi}{4} s_p^2} \times \frac{A}{s \times s_p}$$

where Co = number of corners, P = perimeter, l = longest Feret diameter, A = area, s = shortest Feret diameter, s_p = Feret diameter perpendicular to the shortest Feret diameter. A value of SF below 4.5 indicates a round or elliptic shape, whereas values between 7 and 8 are characteristic for a parallelogram like shape. Fig. 1 illustrates the particle shape of the powders. It can be seen, that if both the values for AR and SF are considered, it is possible to completely describe the shape of the powders chosen.

Figs. 2 and 3 illustrate the changes in the angle of internal friction, δ, and Jenike's flow factor, *ff*, as functions of the magnesium stearate concentration. Table 2 summarizes the results obtained using the annular shear cell for the unlubricated powders in comparison to the mixtures with magnesium stearate, for which the best flow properties had been found, thus for a magnesium stearate concentration which gave the lowest value for δ or the highest value for *ff* (compare Figs. 2 and 3). The angles of internal friction (δ) are presented as the average value obtained from 3 different yield loci for different consolidation loads, because the values were similar. This finding is in agreement with literature reports (Irono and Pilpel, 1982). The addition of magnesium stearate in all cases improved the flow properties of the unlubricated powder, represented by Jenike's flow factor (*ff*), and reduced the angle of internal friction, δ. However, the extent of this effect varied from powder to powder. The largest reduction in δ was obtained for calcium carbonate, and based on the value of *ff*, the powder flow changed from a 'cohesive' into a 'ready flowing' powder. On the other hand, the addition of magnesium stearate to potassium chloride improved the powder flow only slightly without changing the flow category 'ready flowing', and δ also hardly changed. Furthermore, slight changes in δ were not always complemented by only slight changes in *ff*. For example, for microcrystalline cellulose the decrease in the value of δ was only 2°, but the value of *ff* changed to a great extent, indicating the alteration of an originally 'cohesive' into a 'freely flowing' powder. Such discrepancies were previously reported (Jolliffe and Newton, 1982), and indicate that δ and *ff* provide different bulk characteristics of a powder.

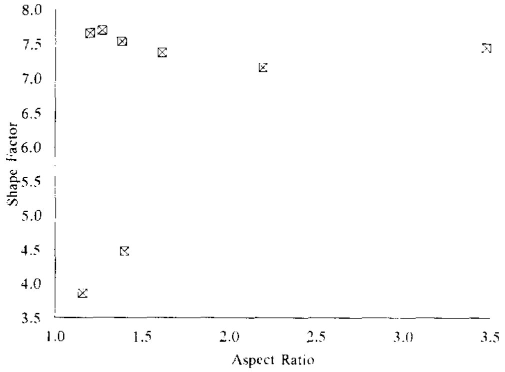

Fig. 1. Shape characteristics for powders.

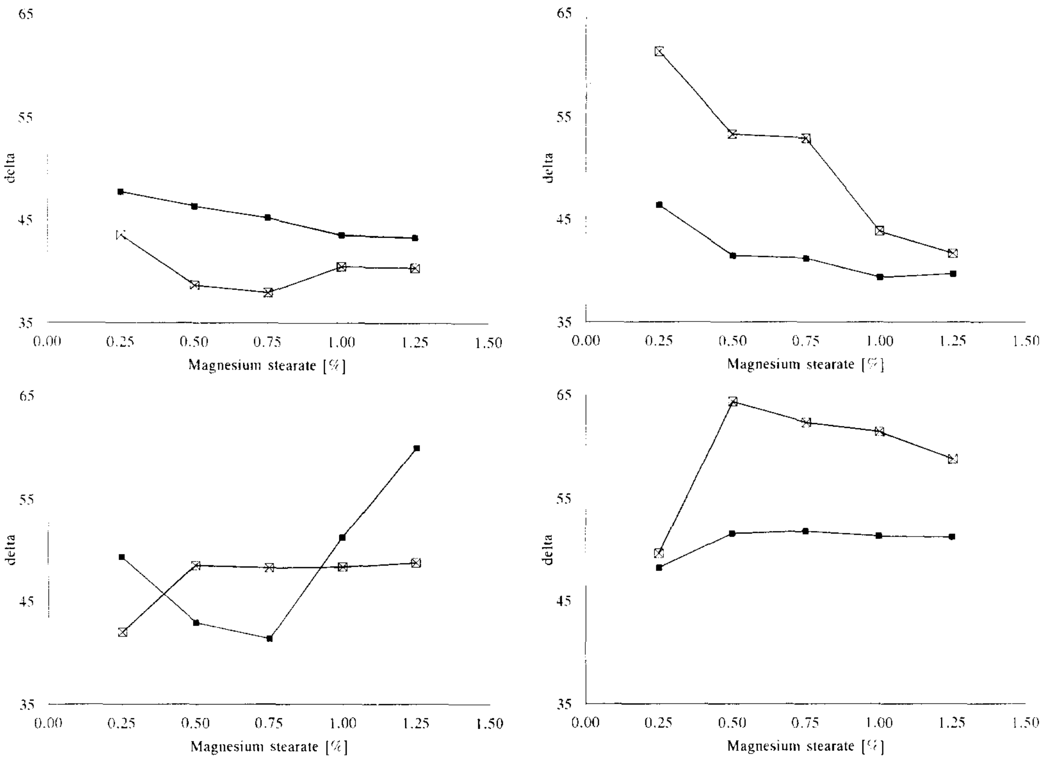

Fig. 2. a–d Angle of internal flow, δ, as a function of the magnesium stearate concentration. 2a: ■, Starch 1500; ☒, paracetamol; 2b: ■, calcium carbonate; ☒, potassium chloride; 2c: ■, maize starch; ☒, microfine cellulose; 2d: ■, microcrystalline cellulose; ☒, acetylsalicylic acid.

At an optimal lubricant concentration, the friction occurring is only partially due to shearing of the lubricant film, but mainly due to deformation of the surface (Bowden and Tabor, 1973). More elastic, bouncing surfaces give less friction. Whether this finding, obtained on engineering surfaces, can be adapted for pharmaceutical materials can be tested using materials of similar origin, e.g. microcrystalline and microfine cellulose or pregelatinized and maize starch. Although similar in origin, the two products in each group are different in their elastic properties, which can be quantified by their Young's modulus of elasticity, E. The value of E is 10.3 GPa and 8.69 GPa for microcrystalline and microfine cellulose, respectively (Bin Baie et al., 1996), and 5.02 GPa (Bin Baie et al., 1996) and 3.71 GPa (Bassam et al., 1990) for pregelatinized and maize starch, respectively. Comparing the values of $\delta_{\min}$ (see Table 2), it can be seen that the higher the friction is, the larger E is. Therefore, Bowden and Tabor's findings are principally adaptable for pharmaceutical materials. However, while for the two cellulose products the change in E by about 1.5 GPa led to a change in more then 6° for $\delta_{\min}$, a similar change in E was accomplished only by a change of 2° for $\delta_{\min}$ with respect to the starch products. Hence, in addition to E, particulate properties need to be considered.

To evaluate the influence of particle size and shape on the flow properties, statistical analysis was applied to the data. A combination of analysis of variance (ANOVA) and regression analysis was used. In the final regression solutions, only influence factors, for which ANOVA had proved their significance, are included. The root mean square deviation (RMS) between predicted and observed data was used as a measure of data fit (residual analysis).

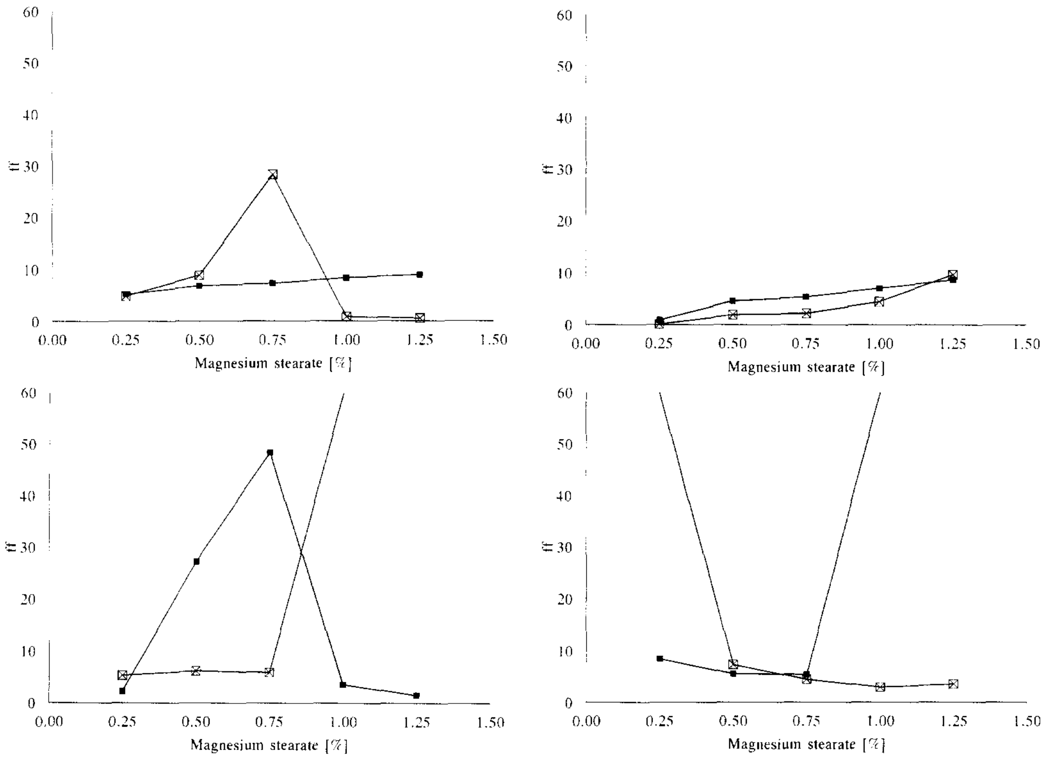

Fig. 3. a–d Jenike's flow factor, *ff*, as a function of the magnesium stearate concentration. 2a: ■, Starch 1500; ☒, paracetamol; 2b: ■, calcium carbonate; ☒, potassium chloride; 2c: ■, maize starch; ☒, microfine cellulose; 2d: ■, microcrystalline cellulose; ☒, acetylsalicylic acid.

The angle of internal friction of the unlubricated powders, δ, is dependent both on particle size and shape and can be well predicted (RMS = 5.2%):

$$\delta = 0.347\cdot e^{AR} - 2.434\cdot \ln d_{vs} + 59.336 \qquad (1)$$

The inclusion of AR as shape factor into the equation suggests that asymmetrical shapes are more prone to cause friction and flow problems than symmetrical shapes. An increase in particle size decreases the value of δ, but this effect is more pronounced in the fine particle size range. Tan and Newton (1990) had also found that for rather symmetrical particles δ decreased with an increase in particle size, more pronounced between the fine and medium size fractions than between a medium and a coarse particle size fraction. However, for the fibrous Avicel®PH101, an increase of δ with increased size was observed.

After addition of the optimal lubricant amount, only the particle shape remains important, whereas the influence of particle size on δ_{min} has become insignificant (RMS = 5.7%):

$$\delta_{min} = -16.312\cdot\frac{1}{AR} + 53.710 \qquad (2)$$

Fig. 4 illustrates the relationship found. The values of AR are incorporated into the equation as their reciprocals, and considering the negative sign of the related regression coefficient, again asymmetry or elongation can be related to increased friction. The outlying point belongs to paracetamol and indicates that there might be further factors which have not been controlled during the study. For the optimal magnesium stearate concentration with respect to $\delta(\rightarrow\delta_{min})$,

Table 2
Friction and flow characteristics of powders

Powder	Unlubricated powder		Lubricated powder			
	δ [°]	*ff*	δ_{min} [°]	$MgSt_{\delta}$ [%]	ff_{max}	$MgSt_{ff}$ [%]
Pregelatinized starch	55.41	2.27	43.30	1.25	8.95	1.25
Paracetamol	45.00	8.78	37.95	0.75	28.27	0.75
Calcium carbonate	56.23	1.81	39.35	1.00	8.67	1.25
Potassium chloride	43.77	5.75	41.67	1.25	9.67	1.25
Maize starch	48.36	9.68	41.35	0.75	48.45	0.75
Microfine cellulose	46.67	9.20	41.99	0.25	>50	1.25
Microcryst. cellulose	50.43	7.66	48.21	0.25	>50	1.25
Acetylsalicylic acid	54.48	1.58	49.61	0.25	>50	0.25

MgSt, optimum magnesium stearate concentration.

no equation which could be used to precisely predict the optimal lubricant amount could be found. However, ANOVA indicated the significance of *AR* and *SF* on this property ($p = 0.006$ an 0.012 for *AR* and *SF*, respectively). Hence both the geometric shape and the asymmetry and elongation of the particles are responsible for a variable optimum lubricant concentration. Variations in shape cause variations in surface area for particles of otherwise similar size, and hence the film thickness of the lubricant differs at the same lubricant concentration for various particle shapes.

For the flow properties of the powders represented by the values of *ff* and ff_{max}, as well as the optimal glidant concentration for $ff(\rightarrow ff_{max})$ no mathematical equation could be found, which was able to predict these properties accurately. However, ANOVA indicated that for the parameters *ff* and ff_{max} only the particle shape was a significant influence factor (*ff*: $p = 0.039$ for *SF*; ff_{max}: $p = 0.001$ for both *AR* and *SF*). This can again be contributed to the surface area available for the glidant film. On the other hand, the optimal glidant concentration appeared to depend only on the particle size ($p = 0.015$). However, from Fig. 5 it can be seen, that over a wide range of size the optimal lubricant concentration is similar. Only for very large particle sizes, less glidant is needed. Large particles are very often already freely flowing without any glidant, and therefore the addi-

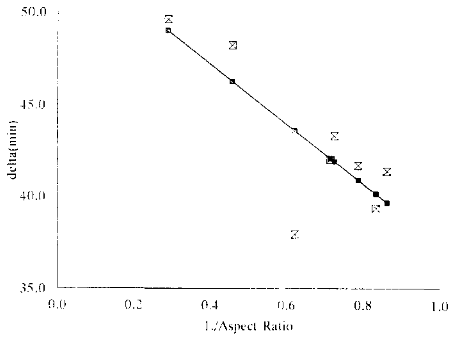

Fig. 4. The influence of particle shape on the angle of internal friction at optimum magnesium stearate concentration. ■, Estimated values; ☒, experimental values.

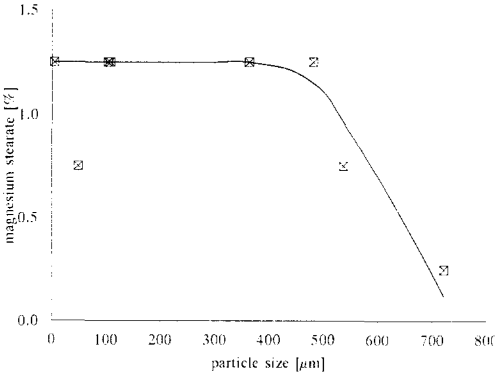

Fig. 5. The influence of particle size on the optimum magnesium stearate concentration for the flow factor. ☒, Experimental values; ——, mathematical function describing the relationship between particle size and magnesium stearate concentration.

tion of a glidant will not lead to a major improvement of powder flow.

It can be concluded, that particle size and in particular particle shape influence the friction and flow properties of powders. While the friction properties depend more on the asymmetry or elongation of the particles, powder flow depends more on the geometric shape. For unlubricated powders, the flow factor increases from needle shape, cubic, angular to round particles. However, at an optimal glidant concentration needle shaped particles can provide similar flow properties to round particles, because the surface area of such particles is less than that of for e.g angular shaped particles.

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